

THE ROLE OF DIFFERENT TRANSPORT MODES ON URBAN PM₁₀ LEVELS IN BUCHAREST AND SZEGED, CENTRAL EUROPE

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REZUMAT. Scopul acestei lucrări este de a identifica principalele zonele geografice responsabile pentru creșterea nivelului de PM₁₀ în două orașe (București, Europa de Est și Szeget Europa Centrală și de Est). Traectoriile înapoi (modelul HYSPLIT) sunt cluster folosind metrica Mahalanobis, în scopul de a determina care regiuni au concentrații ridicate de PM₁₀. În această lucrare au fost analizate valorile mediei zilnice de PM₁₀, precum și datele zilnice meteorologice luate din ambele orașe pe o perioadă de cinci ani (2004-2008).

Cuvinte cheie: calitatea aerului, traectorii înapoi, date meteorologice, PM₁₀.

ABSTRACT. The aim of this paper is to identify the key geographical regions responsible for increase particulate matter (PM₁₀) levels in two cities (Bucharest, Eastern Europe; and Szeged, East-Central Europe). Backward trajectories (HYSPLIT model) are clustered using the Mahalanobis metric in order to determine which regions imply high PM₁₀ concentrations. In this paper was analyzed daily mean PM₁₀ data as well as daily meteorological data taken for both cities for a five years period (2004-2008).

Keywords: air quality, backward trajectories, meteorological data, PM₁₀.

1. INTRODUCTION

PM₁₀ is a measure of particles in the atmosphere with a diameter of less than or equal to a nominal 10 micrometers. The 24-h limit value for PM₁₀ (50 $\mu\text{g}\cdot\text{m}^{-3}$) is frequently exceeded in the urban environment. The short- and long-term human exposure to high particulate matter concentrations observed in urban environment increases the risk of respiratory and cardiovascular (Feng and Yang, 2012) diseases. For Bucharest, the predicted average gain in life expectancy for people of 30 years of age for a decrease in average annual PM_{2.5} level from 38.2 $\mu\text{g}\cdot\text{m}^{-3}$ (2004-2006) to 10 $\mu\text{g}\cdot\text{m}^{-3}$ (World Health Organization, 2000) is 22.1 months (Medina et al., 2004). For Szeged, PM₁₀ counts a major pollutant influencing respiratory diseases (Matyasovszky et al., 2011); furthermore, a set of explanatory variables including PM₁₀ indicates a strong association with allergic asthma emergency room visits (Makra et al., 2012). Therefore, studying potential key regions and long-range transport effects on urban PM₁₀ levels is of great importance.

The aim of this paper is to identify the key geographical regions responsible for PM₁₀ levels in two cities (Bucharest, Eastern Europe; and Szeged, East-Central Europe). Backward trajectories arriving

at these sites are clustered using the Mahalanobis metric in order to determine which regions imply high PM₁₀ concentrations. The clustering is performed using three-dimensional (3D) backward trajectories. ANOVA is used to determine whether PM₁₀ concentrations corresponding to these trajectory clusters differ significantly. Cluster-dependent occurrences, when 24-h mean PM₁₀ concentrations exceed the limit value of 50 $\mu\text{g}\cdot\text{m}^{-3}$ are also analyzed with two statistical indices. Lastly, a statistical procedure is developed in order to separate medium-range PM₁₀ transport including local PM₁₀ emissions from the long-range transport of PM₁₀.

2. DATA AND METHODS

2.1 Study areas and monitoring data

Five years (2004-2008) of daily mean PM₁₀ data as well as daily meteorological data (mean temperature, mean global solar flux, mean relative humidity and daily precipitation total) taken from two European cities – Bucharest (Romania) and Szeged (Hungary) – were analyzed. The reasons for selecting these sites include their fairly big distance (710 km) and their substantial difference in topography and climate. Namely, Bucharest ($\varphi = 44.43^\circ\text{N}$;

$\lambda = 26.10^{\circ}\text{E}$; $h = 74$ m a.s.l.), the capital of Romania, is located in the southeast of the country. The city lies on the banks of the Dâmbovița River, about 70 km north of the Danube. Szeged ($\varphi = 46.25^{\circ}\text{N}$; $\lambda = 20.10^{\circ}\text{E}$; $h = 79$ m a.s.l.), the largest settlement in SE Hungary, is located at the confluence of the Rivers Tisza and Maros.

2.2. Backward trajectories

In the frame of an ETEX (European Tracer Experiment) research, efficacy of three large-scale Lagrangian dispersion models (CALPUFF 5.8, FLEXPART 6.2 and HYSPLIT 4.8) was compared. As the HYSPLIT model has the best performance according to four statistical scores (Anderson, 2008) we decided to use the HYSPLIT model (Draxler and Hess, 1998).

Backward trajectories for Szeged and Bucharest corresponding to the Hybrid Single-Particle Lagrangian Integral Trajectory (HYSPLIT, version 4.8; <http://www.arl.noaa.gov/ready/hysplit4.html>) model (Draxler and Hess, 1998) were obtained from National Centres for Environmental Prediction / National Centre for Atmospheric Research (NCEP/NCAR; <http://dss.ucar.edu/datasets/ds090.0/>).

Since a single backward trajectory has a large uncertainty and is of limited significance (Stohl, 1998), a three-dimensional (3D) representation of the synoptic air currents in the given regions was made via the reconstruction and analysis of a large number of atmospheric trajectories. 4-day, 6-h 3D backward trajectories arriving at the two locations at 1200 Greenwich Mean Time (GMT) at heights $h=500$, 1500 and 3000 m above ground level (a.g.l.) for each day over a 5-year period from 2004 to 2008 were taken in order to describe the horizontal and vertical movements of an air parcel for the above-mentioned two cities. These three arrival heights were selected in order to understand the behaviour of the air masses circulating in the boundary layer (BL) and the free troposphere (FT): 500 m (typical for the near surface), 1500 m (representative for the BL top) and 3000 m (characteristic for the FT heights) (Córdoba-Jabonero et al., 2011). The actual heights of trajectories may act as an indicator of the atmosphere-surface interactions. For instance, an air mass moving over a source area at low vertical levels might be more affected by PM10 loadings of this region than another air mass travelling at much higher levels over this same area.

2.3. Cluster analysis

Cluster analysis classifies trajectories with similar paths. The aim of any clustering technique is

to maximize the homogeneity of elements (in our case, backward trajectories) within the clusters and also to maximize the heterogeneity among the clusters. Here a non-hierarchical cluster analysis with the k-means method (Anderberg, 1973) was applied using the Mahalanobis metric (Mahalanobis, 1936) available in MATLAB 7.5.0. Input data as clustering variables include the 6-hourly co-ordinate values (φ - latitude, λ - longitude and h - height above ground level (a.g.l.) of the 4-day 3D backtrajectories for both cities and the three given heights.

The homogeneity within clusters was measured by RMSD defined as the sum of the root mean square deviations of cluster elements from the corresponding cluster centre over clusters. As the RMSD will usually decrease with an increasing number of clusters this quantity is not very useful for deciding about the optimal number of clusters. However, the change of RMSD (CRMSD) versus the change of cluster numbers, or rather the change of CMRSD (CCRMSD) is much more informative. Here, working with cluster numbers from 15 to 1, an optimal cluster number was selected so as to maximize the change in CRMSD. The rationale behind this approach is that the number of clusters producing the largest improvement in cluster performance compared to that for a smaller number of clusters is considered optimal.

The results of our cluster analysis are discussed and presented only for the lowest ($h=500$ m a.g.l.) arrival height because backtrajectories at this arrival height are expected to have the largest influence on the PM10 concentration of the target site. The separation of the backward trajectory clusters and preparation of figures for clusters of backward trajectories were performed using a novel approach that employs a function called "convhull". The algorithm (qhull procedure; <http://www.qhull.org>) gathers the extreme trajectory positions (positions farthest from the centre) belonging to a cluster, which are then enclosed. Specifically, the procedure creates the smallest convex hull with minimum volume covering the backtrajectories of the clusters (Preparata and Hong, 1977).

Trajectory clusters are projected on a stereographic polar plane supported by HYSPLIT (Taylor, 1997).

2.4. Analysis of variance (ANOVA)

ANOVA is used to test whether the means of PM10 values under different trajectory types (clusters) differ significantly for a given city. If ANOVA, based on the F-test, detects significant difference among these means another test is then

applied to determine which means differ significantly from the others. Significant differences among mean PM10 concentrations under different trajectory types may tell us about the origin and transport of air masses on local PM10 levels. There are several versions available for comparing means calculated from subsamples of a sample. A relatively simple but effective way is to use the Tukey test. It performs well in terms of both the accumulation of first order errors of the test and the test power (Tukey, 1985).

ANOVA assumes in general that elements of the entire data set represented as random variables are independent, and elements within each group have identical probability distributions. Daily PM10 data, however, do not meet these requirements as they have an annual trend in both the expected value and variance. These trends can be removed by standardization. Standardized data are free of annual trends and thus distinguishing between average PM10 levels corresponding to trajectory types is due to the types themselves and it is not related to periods of the year. (Note that the standardized PM10 values are dimensionless.) The annual trend of the expected value is estimated by fitting sine and cosine waves with periods of one year and half a year to PM10 data by the least squares technique. Note that the half a year period is introduced to describe the temporal asymmetry of the annual trend. A subtraction of the estimated trend from data results in centralized data. The annual trend of the variance is estimated by fitting sine and cosine waves with periods of one year and half a year to squared centralized data. Lastly, centralized data are divided by the root of the estimated time-dependent variance in order to get standardized data. Consecutive daily PM10 values are correlated and produce higher variances of the means estimated under trajectory types compared to those for uncorrelated data. The autocorrelation structure is modeled via first order autoregressive (AR) processes conditioned on clusters. The classical Tukey-test (Tukey, 1985) is then modified according to the variances of estimated means obtained with the help of the AR models. Note that the omission of this step could systematically overestimate the number of significantly different means.

2.5. Factor analysis and special transformation

Factor analysis (FA) identifies linear relationships among subsets of examined variables, which helps to reduce the dimensionality of the initial database without substantial loss of information. First, a factor analysis was applied to the initial standardized

dataset consisting of 12 variables (3 climatic and 9 trajectory variables introduced in Section 4) in order to reduce the original set of variables to fewer variables. These new variables called factors can be viewed as the main climate/trajectory features that potentially influence the daily mean PM10 concentration. The optimum number of retained factors is determined by the criterion of reaching a pre-specified percentage of the total variance (Jolliffe, 1993). This percentage value was set at 80% in our case. Next, a further data manipulation on the retained factors called special transformation (Jahn and Vahle, 1968) was performed to discover to what degree the above-mentioned explanatory variables (3 climatic and 9 trajectory variables) affect the resultant variable (daily mean PM10 concentration), and to give a rank of their importance.

2.6. Statistical characterization of PM10 exceedance episodes

The role of long-range transport is studied by analyzing the cluster occurrences on days when 24-h mean PM10 concentrations exceed the limit value of $50 \mu\text{g}\cdot\text{m}^{-3}$. Two statistical indices of daily PM10 exceedance episodes associated with trajectory clusters are calculated in the same manner as in Borge et al. (2007). For a given site and cluster i , INDEX1 is defined as

$$INDEX1_i(\%) = \frac{D_{(>50)_i} \cdot 100}{D_i}, \quad (1)$$

where: D_i is the number of occurrences of cluster i ; $D_{(>50)_i}$ – the number of 24-h PM10 exceedances. INDEX1 gives the likelihood of an exceedance for a given cluster. INDEX2 is defined as

$$INDEX2_i(\%) = \frac{D_{(>50)_i} \cdot 100}{E}, \quad (2)$$

where: E is the total number of 24-h PM10 exceedance days recorded at a given site. INDEX2 can be interpreted as the likelihood of certain trajectory being present on a PM10 exceedance day.

3. RESULTS

3.1. Bucharest

The 3D clustering produced eleven clusters based on CCRMSD. All of the trajectories with colour-coded clusters, all of the clusters without trajectories but with their 3D convex hulls for the top view, in addition mean backward trajectories of the clusters for the top view, and all trajectory clusters enclosed

by their transparent 3D convex hull as well as their 90° rotated version are presented in Fig. 1. A vertical view of the trajectory clusters enclosed by their transparent 3D convex hulls has also been added (Fig. 2). Pairwise comparisons of the cluster averages found 5 significant differences among the possible 55 cluster pairs (9.1%). Hereafter only clusters of the above-mentioned significantly different cluster-averaged PM10 levels were then considered and analyzed (Fig. 2).

Clusters 5 and 6 have the highest INDEX1 values, namely 70.1% and 74.2%, respectively (Fig. 5). The high INDEX1 values of these clusters (Fig. 5) are in agreement with their high mean PM10 concentrations. The high standard deviation corresponding

to cluster 5 implies a higher chance of extreme PM10 episodes for this cluster. Note that INDEX1 and INDEX2 are not independent parameters. When a cluster is frequent, a high INDEX1 value involves a high INDEX2 value (see cluster 5) (Fig. 5). The highest frequency of cluster 5 and a relatively high occurrence of cluster 6 (having the 4th highest frequency of the clusters) emphasize the importance of these clusters in PM10 transport. The low-moving backward trajectories of cluster 6 further raise the significance of this cluster in long-range transport. Cluster 5 comprises high-moving backtrajectories that weakens the role in transporting particulates (Fig. 1, upper left panel, middle right panel, as well as the lower left and right panels; Fig. 2).

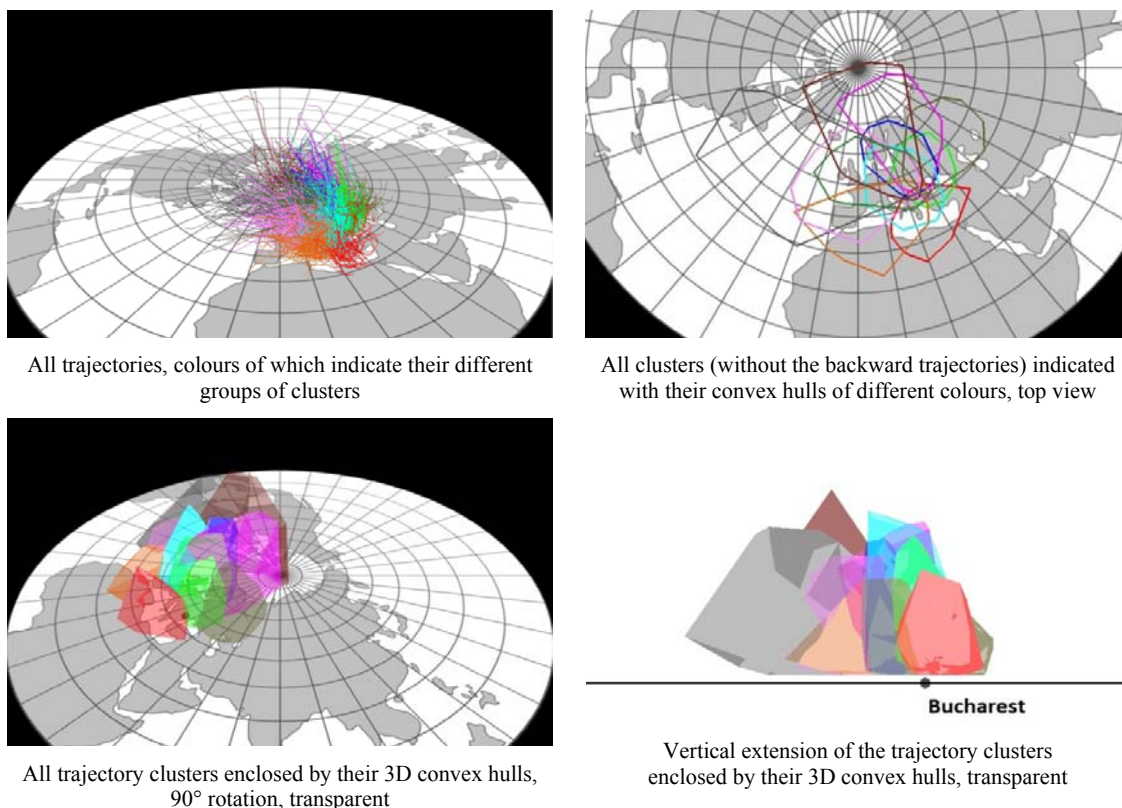


Fig. 1. 3D clusters of backward trajectories retained, Bucharest, $h = 500$ m.

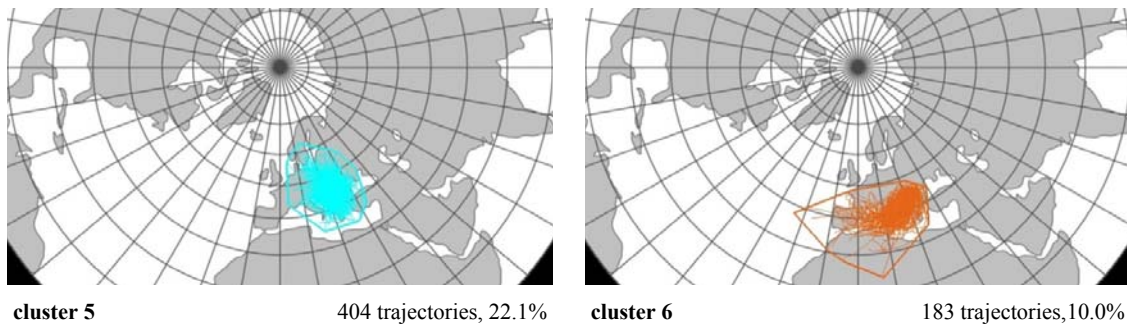
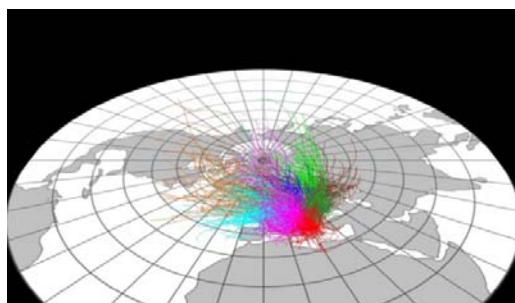


Fig. 2. The individual clusters of the backward trajectories retained, enclosed by their convex hulls, Bucharest, top view, $h = 500$ m.

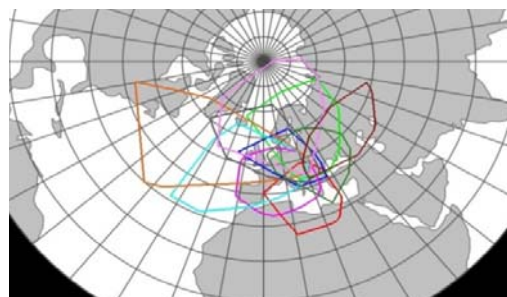
3.2 Szeged

Ten clusters were retained in a 3D analysis based on CCRMSD (Figs. 3 and 4). The individual clusters (Fig. 4) with the name of the source areas and their standardised average PM10 levels are presented. For Szeged, 20 significant differences were detected among the possible 45 cluster pairs (44.4%).

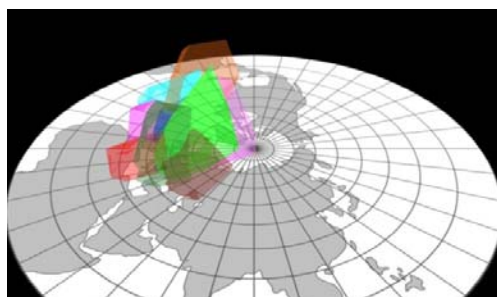
The highest INDEX1 value (57.3%) is associated with cluster 1 (Southern Europe with North Africa) with relatively low frequency (7.2%) (Fig. 5). The next highest INDEX1 values, in decreasing order, belong to cluster 10 (Eastern Europe with regions over the West Siberian Plain) (38.4%) and cluster 9 (Central Europe) (37.8%). This is in agreement with the fact that these clusters have high mean PM10 levels (Fig. 5).



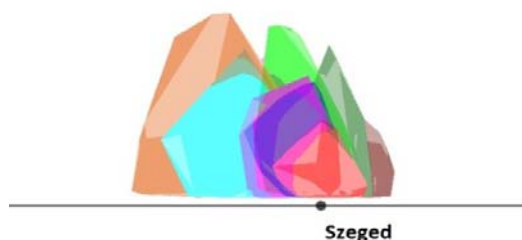
All trajectories, colours of which indicate their different groups of clusters



All clusters (without the backward trajectories) indicated with their convex hulls of different colours, top view

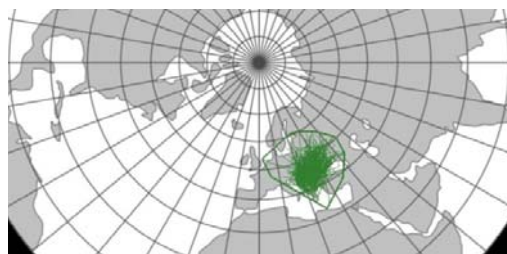


All trajectory clusters enclosed by their 3D convex hulls, 90° rotation, transparent



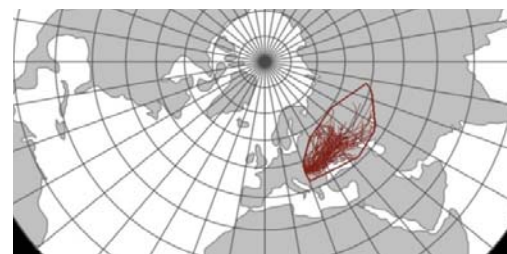
Vertical extension of the trajectory clusters enclosed by their 3D convex hulls, transparent

Fig. 3. 3D clusters of the backward trajectories retained, Szeged, $h = 500$ m.



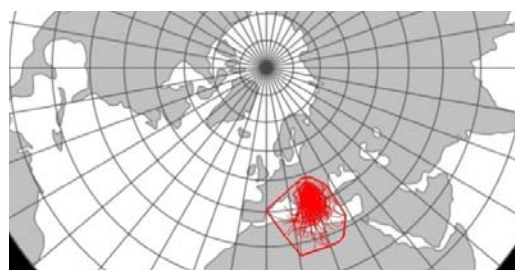
cluster 9

280 trajectories, 15.3%



cluster 10

73 trajectories, 4.0%



cluster 1

132 trajectories, 7.2%

Fig. 4. The individual clusters of the backward trajectories retained, enclosed by their convex hulls, Szeged, top view, $h = 500$ m

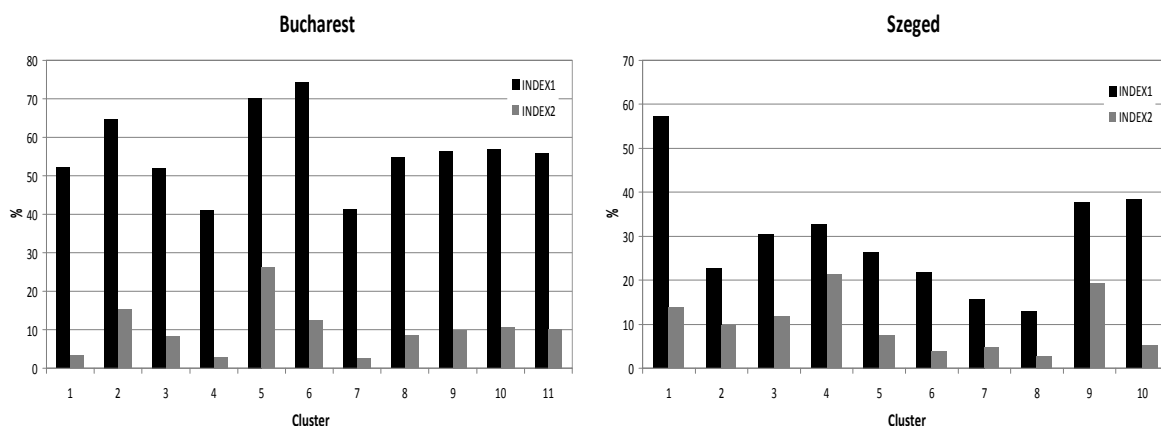


Fig. 5. Indices 1 and 2 for 3D clusters of the backward trajectories, $h = 500$ m.

4. DISCUSSION AND CONCLUSIONS

A cluster analysis was applied to 4-day, 6-hourly backward trajectories arriving at Bucharest and Szeged over a 5-year period in order to identify the main atmospheric circulation pathways influencing PM10 levels at these sites. When performing ANOVA, the decision on the significance of two cluster averages is based on a modified *t*-test because the test is performed using standardized data instead of the original data. The Mahalanobis metric was used in order to avoid the need for a two-stage cluster analysis introduced in Borge et al. (2007). The 3D delimitation of the clusters by the function “convhull” is a novel approach. Furthermore, presentation of vertical extension of the trajectory clusters enclosed by their 3D convex hulls and, in this way, delimiting low-moving backtrajectories is a novel procedure. Furthermore, no papers have been published so far studying PM10 transport for Eastern European target stations using backward trajectories.

After classifying objective groups of backtrajectories and, in this way, detecting the main circulation pathways for the cities in question, it is important to separate local and transported components of the actual PM10 levels. In other words, it is necessary to determine the relative weight of these two components in the measured PM10 concentration. There are several case studies available that allow one to distinguish the long-range PM10 transport episodes from local PM10 pollution episodes (Aarnio et al., 2008). Masiol et al. (2012) applied a chemometric analysis and a source apportionment model for discriminating local processes and long-range transport on particulate matter levels. Wong et al. (2013) discerned short- and long distance sources on the types of aerosols. Juda-Rezler et al. (2011) developed a combination of different methods to distinguish long-range transport and regional transport from local pollution sources. Analyses of local meteorological

conditions and air-mass backtrajectories for a given city play an important role in developing methods for the above purpose (Aarnio et al., 2008). An attempt is made here to discriminate these two pollution modes (i.e. local PM10 emission and long-range PM10 transport) in the entire 5-year data set using local meteorological parameters and components of the backtrajectories. Local PM10 pollution is characterized next via the daily mean temperature, daily relative humidity and daily global solar flux. Long-range PM10 transport is described by (1) the real 3D length of the backtrajectories, (2) the length of the 3D backtrajectories as the crow flies, (3) their ratio, (4) the average daily highest and (5) lowest positions of the backward trajectories based on their 4-day, 6-hourly positions. In order to take into account further characteristics of long-range PM10 transport, stereographic plane projection of each backtrajectory is considered. The target station is located into the origin of an imaginary frame of reference. Further parameters of the long-range transport are as follows: *x* coordinates belonging to the (6) easternmost and the (7) westernmost points of the given backtrajectory, as well as the *y* coordinates belonging to the (8) northernmost and the (9) southernmost points of the same given backtrajectory. The average daily highest and lowest positions of the backward trajectories refer to the vertical transport of PM10 in the atmosphere, which comes from either turbulent transport dominating the vertical exchange of PM10 in the boundary layer or intense convective upwelling, which results in large amounts of particulates being transported from near the surface to high elevations. The latter four (6-9) characteristics represent the extreme points of a backward trajectory both to east-west and north-south directions on a horizontal plane, representing the east-west and north-south extension of the long-range transport.

As the PM10 level on a given day is substantially influenced by whether conditions such as precipita-

tion, the backward trajectories are divided into two groups, i.e. non-rainy and rainy days of the arriving sites. This kind of classification of days reveals the role of precipitation in the quantity of transported PM10 (Querol et al., 2009). Factor analysis with special transformation was carried out for both cities with the two groups (rainy or non-rainy days) and the 500 m, 1500 m and 3000 m arrival heights of the backward trajectories, separately. Thus, altogether $2 \times 2 \times 3 = 12$ procedures gave information about the weights of the local source and long-range transport reflected by the 12 explanatory variables. The main conclusions are as follows.

Considering the 500 m arrival height, long-range PM10 transport plays a higher role compared to local PM10 emission both for non-rainy and rainy days for Bucharest and also for Szeged. Predominance of long-range transport compared to local emission is higher in Bucharest than in Szeged on non-rainy days, while it is equally higher for both cities on rainy days. As regards the components of the two different transport modes on non-rainy days, the local variables are equally important for both cities and all three heights. The components associated to the length of the backward trajectories have equally high weights for both cities, furthermore their east-west components have also substantial role for both cities and all three heights. In addition, the role of the north-south components is more important for Szeged. For rainy days, components of neither the local nor the long-range transport are important for Bucharest, while temperature and global solar flux as well as the east-west components of the long range transport are the most relevant for Szeged. Adding up the weights of the local pollution and long-range transport, the average value of the two weights is larger for both Bucharest and Szeged on non-rainy days and for Bucharest on rainy days at 500 m height compared to the higher levels. Hence, the twelve variables contain more information on PM10 when using backtrajectories arriving at 500 m height in these three cases. For the remaining case (Szeged on rainy days) the results (the average weight of the local pollution and long-range transport is the lowest at 500 height compared to the remaining two arrival heights) disagree with our preliminary expectations because near surface air currents might be affected by several factors that substantially modify the ratio of the local and transported particulates. Moreover, the variables contain more information at higher levels on the transported PM10 for Szeged on rainy days.

In a subsequent examination, another factor analysis with special transformation was performed for rainy and non-rainy days including collectively the 500 m, 1500 m and 3000 m arrival heights of the

backward trajectories. In this way, altogether $2 \times 2 = 4$ procedures were implemented. Each procedure comprised 30 explanatory variables in all (3 parameters gave information about the weights of the local source, while the long-range transport was reflected by $3 \times 9 = 27$ explanatory variables, due to the three arrival heights of the backtrajectories). The major results are as follows.

For non-rainy days, components of the long-range transport for the 500 m arrival height have the most important role in determining PM10 concentration both for Bucharest and Szeged. Furthermore, global solar flux and relative humidity, as components of the medium-range PM10 transport including local PM10 emission, as well as the east-west components of the long-range transport for all three heights are within the first ten most important explanatory variables for both cities. For rainy days, only real 3D length of the backtrajectories at 3000 m height is in an important association with the PM10 concentration for Bucharest. For Szeged, both temperature and global solar flux have again an important role. In addition, east-west components of the backtrajectories at 500 m height, as well as average daily highest and lowest positions of the backward trajectories at both 1500 m and 3000 m heights are the most relevant explanatory variables.

For both kinds of factor analysis with special transformation, temperature and global solar flux are in significant negative, while relative humidity is in significant positive association with PM10 concentration. These associations assume an anticyclone ridge weather situation, when descending air currents prevent vertical mixing of the polluted urban and, hence, air pollution can accumulate. These situations with cloudy weather involve a decrease of temperature and global solar flux and an increase of relative humidity.

For both cities and all three heights, components of the backtrajectories are directly proportional to the resultant variable. Namely, bigger length as well as more extreme horizontal and vertical components of the backtrajectories involves higher PM10 concentrations.

Note that these findings are valid only for variations of the daily PM10 concentrations accounted for by the explanatory variables and nothing is known about the variance portion not explained by these variables.

Acknowledgements

The authors would like to thank Roland Draxler for his useful advice and consultations on the HYSPLIT model, version 4.8; to Gabriel Ciuiu

(Environmental Protection Agency, Bucharest) and to Gábor Motika (Environmental Protection Inspectorate of Lower-Tisza Region, Szeged) for providing daily PM10 and meteorological data. This study was supported by the TRANSAIRCULTUR project (No. HURO/1001/139/1.3.4) Hungary-Romania Cross-Border Co-operation Programme 2007-2013, under the auspices of the European Union and the European Regional Development Fund.

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